Extent and Severity of Arsenic Contamination in Soils of Bangladesh

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INTRODUCTION

Since arsenic in groundwater was recognized as a potential threat to human life, much effort has been directed towards ensuring safe drinking water either through mitigation techniques or through finding alternative water sources. Even when alternate sources of water are used for potable purposes, continued cropping of long-term irrigated soils subjected to As contaminated ground water will pose significant risk to animal and human health through soil-crop transfer of As. Food crops such as vegetables and cereals can become a path by which As may enter the food chain, because they can reflect the levels of As that exist in the environment in which they are cultivated (soil and irrigation water). Recent studies by Meharg *et al*. (2002) and Huq *et al*. (2001) demonstrate significant uptake of As by rice and a range of vegetable crops commonly grown in Bangladesh. Numerous greenhouse studies by a number of researchers have revealed that an increase in As in cultivated soils leads to an increase in the levels of As in edible vegetables (Burló *et al*., 1999; Carbonell-Barrachina *et al*., 1999; Helgensen and Larsen, 1999) with many complex factors affecting bioavailability, uptake and phytotoxicity of As (Carbonell-Barrachina *et al*., 1999). Although, there are many studies on As in Bangladesh few have focused on soil As. Considering this, a project was initiated among others, to assess the extent and severity of

As contamination of soils at contrasting sites and develop an understanding of the fate and behavior of As in soil. One of the specific objectives of the project was to characterize soils and associated GW for the assessment of the severity and extent of As contamination. Such a study was considered essential for devising remedial measures to minimize adverse impacts of groundwater and soil As on human health. Along with this, the project aimed at estimating background concentration of As in Bangladesh soils, although limited to several thanas considered in this study. Background concentration is defined as the ambient concentration of As in soils without human influence (Gough, 1993) which depicts an idealized situation (Chen *et al*., 2001). Arsenic present in soils irrigated with contaminated ground water is defined as Anthropogenic As.

MATERIALS AND METHODS

Collection of Soil Samples

For this study, information about As contamination was obtained from secondary sources (DPHE/BGS, 1999; DCH, 1999). For sample collection, As contaminated areas were divided into: Gangetic alluvium flood plain, Teesta alluvium floodplain, Meghna Brahmaputra alluvium flood plain and Pleistocene terrace belts. Table 1 shows the areas from where samples were collected. As such soil, water and crop/vegetable samples were collected covering 15 districts, 27 police stations, 40 unions and 73 villages.

Table 1: Name of the districts selected for soil sample collection

Gangetic alluvium floodplain	Teesta alluvium floodplain	Meghna-Brahmaputra alluvium flood plain	Pleistocene Terrace
1. Jessore	1. Rangpur	1. Comilla	1.Dhaka
2. Kushtria	2. Kurigram	2. Narayanganj	2. Dinajpur
3. Meherpur	3. Pabna	3. Munsiganj	
4. Chuadanga		4. Noakhali	
5. Faridpur		5. Brahmonbaria	

A number of strategies were adopted during soil sampling to ensure that the samples were representative of the region most severely affected by As poisoning. Key issues considered were: frequency of poisoning reported, As content of tube well water, other project activities and farming practices. A summary of our approach to sampling is illustrated in Figure 1.

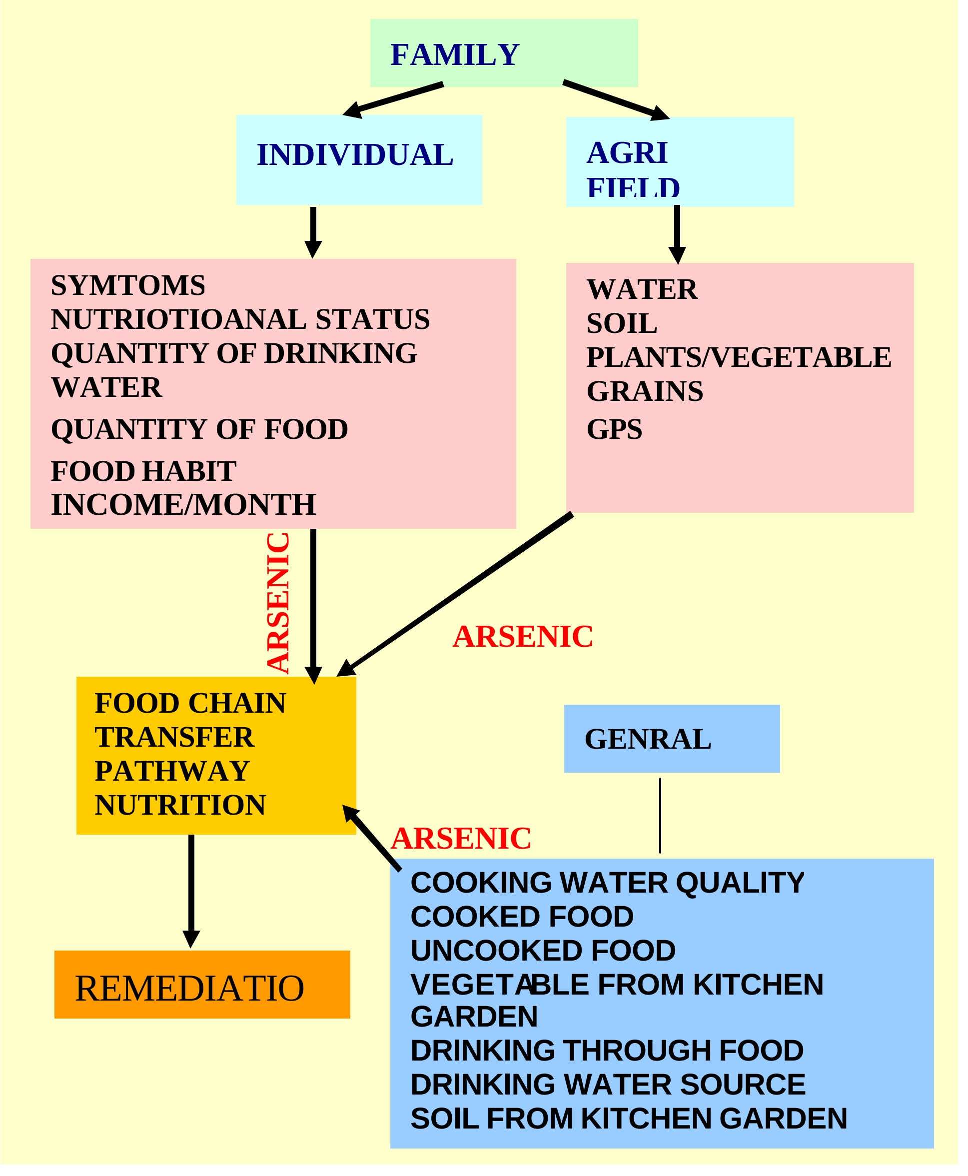

Figure 1: The approach to field sampling

Sampling was restricted to backyard gardens vegetable patch within 100 m of the village and included: tube well(s) water samples, soil samples from the wet patches around the tube well(s), soil samples (0 to 15 cm; 15 to 30 cm) from backyard vegetables garden, growing vegetables/plant samples and soil and crop/plant samples from the agricultural fields nearby. Along with these, samples of irrigation water (shallow tube well), weeds, and fodder were also collected. Here, results of soil samples are presented only. Figure 2a presents a schematic diagram illustrating a typical village setting and Figure 2b shows the sampling sites. The different intensity of shades in the map in Fg. 2b indicate high-risk areas, medium to low risk areas and unaffected areas.

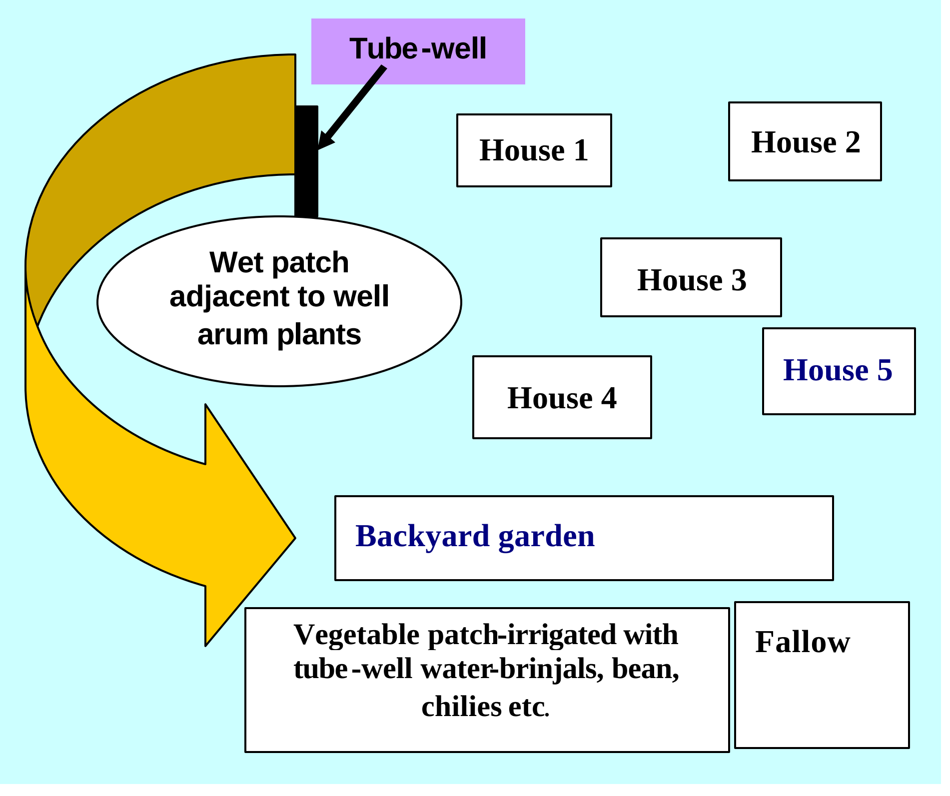

Figure 2a: Schematic diagram illustrating a typical village, tube well and garden activities around residential properties

Water

Midstream water samples (100 ml) were collected from tube-wells or irrigation pumps after initial pumping for 5 minutes. Redox potential of tube well water samples collected was low as indicated by the presence of high concentrations of Fe^{2+} and As^{III} as well as low Eh values that rapidly changed to positive values following exposure to air. Immediately after sampling, 1 ml of concentrated HCl was added to the 100 ml vials containing water and transported to the laboratory for further analysis. Vials were filled to the top and following transfer to the laboratory on ice, the samples were centrifuged at high speed, filtered through 0.45 μ Millipore filters and analyzed for As using HG-AAS.

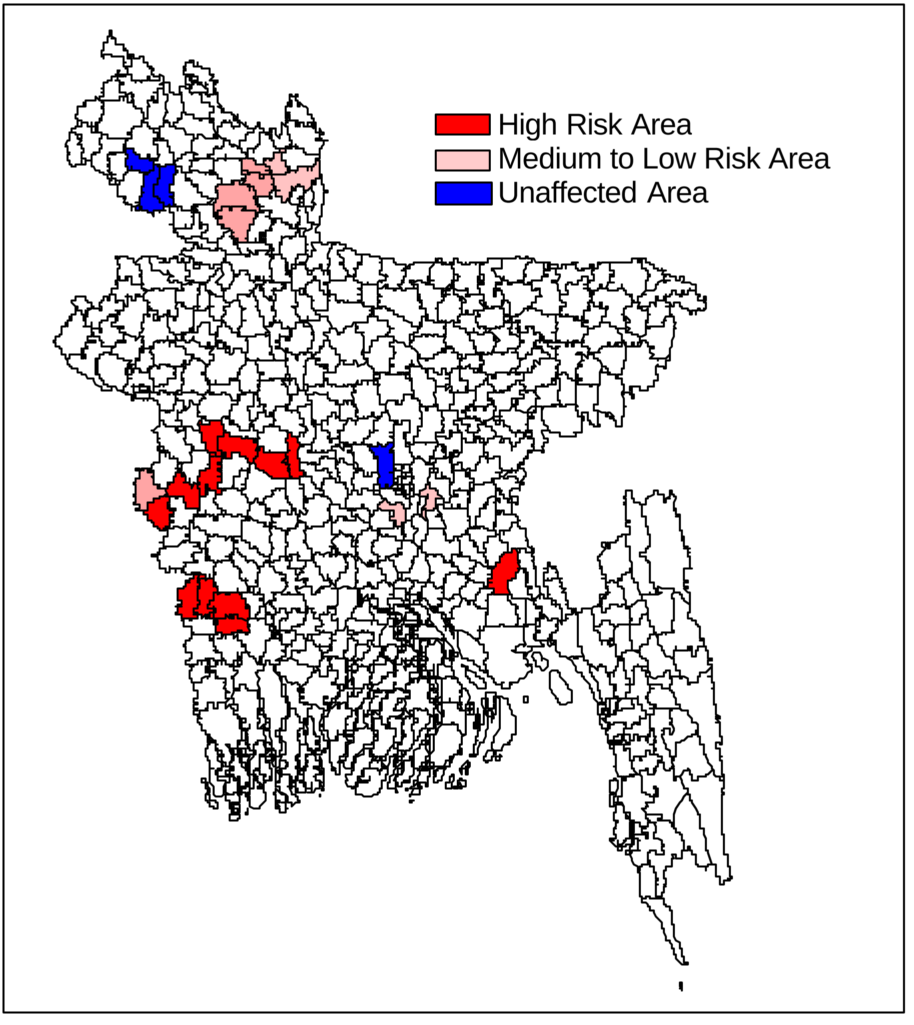

Figure 2b: Sampling sites

Soils

Replicate surface (0-15 cm) and subsurface (15-30 cm) soil samples were collected from each site where water and vegetables were collected and where As poisoning of people has been reported. Each replicate soil sample was a composite of 10 sub-samples (taken to the same depth). In order to monitor the As load on soils from water, samples were collected from regions subjected to hand tube-well, shallow tube-well, deep tube-well and surface water irrigation. Grid sampling was adopted and from each site the number of samples collected ranged from 25 to 40 per acre. After collection, samples were air dried, ground and screened to pass through a 0.5 mm sieve and stored in plastic vials for all laboratory analysis.

Soil Analysis for Arsenic

Aqua-regia extractable As

Extractable Arsenic content of soils was assessed following aqua-regia hot plate digestion of sub samples (0.5 g and 3 replicates) of soils. Soils were digested with three aliquots of aqua-regia (5 ml) solution. Following digestion, the extracts were diluted to 50 mls using aqua regia and As in the extract was estimated by HG-AAS following calibration of the equipment. For every 10-soil sample, we included a certified reference material (CRMs) to ensure QA/QC. Analyses of the extract showed that the digest reproduced 95% of the total As content reported for the CRMs.

Water-soluble As

Soluble As was extracted by shaking samples of soil at a 1:5 soil-water ratios for 24 hours. The determination was the same as that for extractable As.

RESULTS AND DISCUSSION

Figure 3 shows the Arsenic contents in different depths of soils collected from As affected area and As unaffected areas. With one or two exceptions, in most cases, the top 0-150 mm contain more As than the bottom 150-300mm. The average As contents in Bangladesh soils is less than 10 mg/kg. In areas where As contamination of ground water has not been reported, the soil As content is much below the average value.

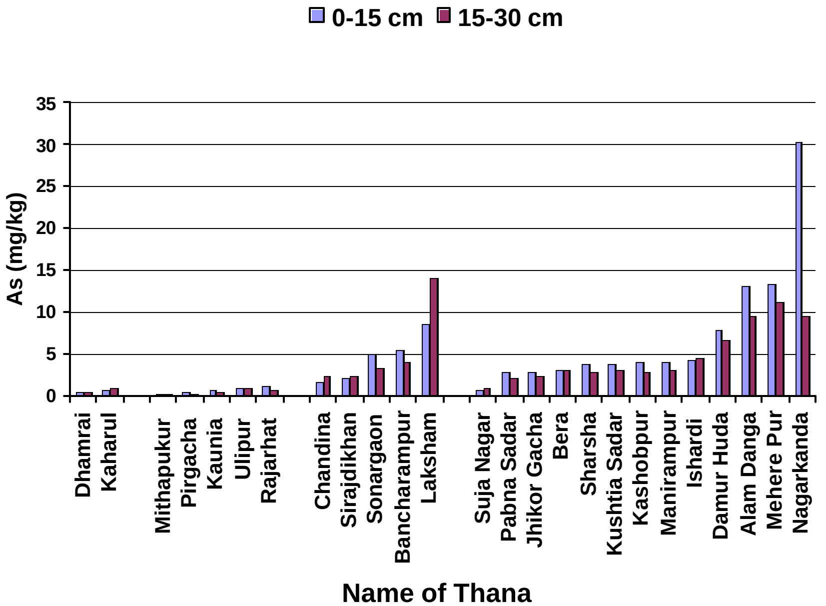

Figure 3: Average soil As values at two depths in different thanas

There is a positively skewed distribution with a long tail towards the right meaning higher values indicating that the concentrations of As in the majority of soils is low with only a few high values; this suggests that the concentrations of As in surface soils are largely not affected by human activities meaning irrigation (Figures 4 and 5).

The current regulatory soil screening levels as well as soil cleanup goals for remediation of contaminated sites vary greatly internationally but are also under developed in many countries. For instance, the regulatory limits established for environment health investigation levels and Human Health investigation levels in Australia are 20 and 100 mg kg^{-1} (NEPC-ACS, 1999). This suggests that in agricultural soils exceeding 20 mg kg^{-1} will require cleanup. In contrast, the regulatory limit established by UK is set at 10 mg kg^{-1} for domestic gardens, and 40 mg kg^{-1} for parks, playing fields and open spaces (O'Neil, 1990).

Much tighter guidelines (0.80 mg kg^{-1} for residential and 3.7 mg kg^{-1} for non residential) have been established in Florida (Tonner-Navarro *et al.*, 1998) based on direct arsenic exposure. Similar variations exist in other countries although in Bangladesh such a guideline does not exist. The average value of < 10mg/kg of As meets the guidelines for residential soils of 100 mg kg^{-1} as required by Australian Health, and 20 mg kg^{-1} as required by the Environmental Guidelines. However, although not in general, in some places, where As contaminated ground water is used for irrigation purpose, the As values are elevated. Values as high as 81 mg/kg have been recorded in surface samples at some sites. The irrigation water that is being used on this soil was found to have As concentration of 0.077mg/L. Nevertheless, there was no significant correlation between As content of irrigation water and surface soil As loading. On the other hand, with much elevated As in irrigation than this, the soil As content was within the average value. In fact, the correlation coefficient values for water-As and soil-As have been found to be negative. This indicates the fact that retention of As by soil is governed by its properties, particularly the nature and amount of clay contents.

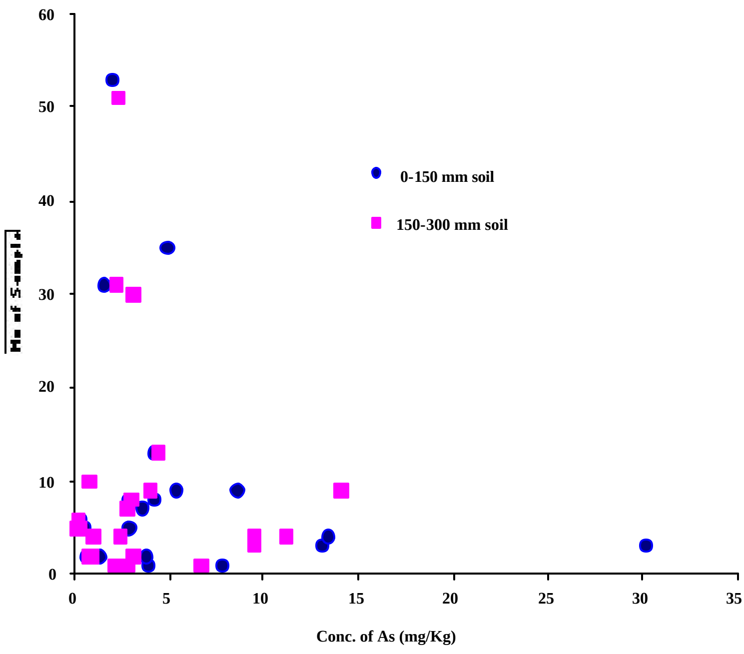

Figure 4: Distribution of arsenic in soil samples

Under laboratory condition both adsorption as well as column leaching studies with soils having different clay contents revealed this. In an adsorption study with three different soils with clay content ranging from

17% to more that 80% clay, it was observed that adsorption of As from solution was greater in higher clay containing soils. Moreover, this adsorption was greater with increasing As concentration in the solution (Fig. 6).

When calculated in terms of kg/ha, the values can be as high 70-75 kg, a value rather high for an undesired element like arsenic (figure-7). On the basis of these observations, it has been estimated that typical soils, even if these have an As-contaminated ground water have an As content of less than 10 kg /ha; soils receiving As contaminated ground water irrigation can have values equal to the average value or higher values while contaminated soils can have values of about 100 kg/ha (figure-8). Soils where the ground water is not contaminated, the As value is lower than 10 kg/ha.

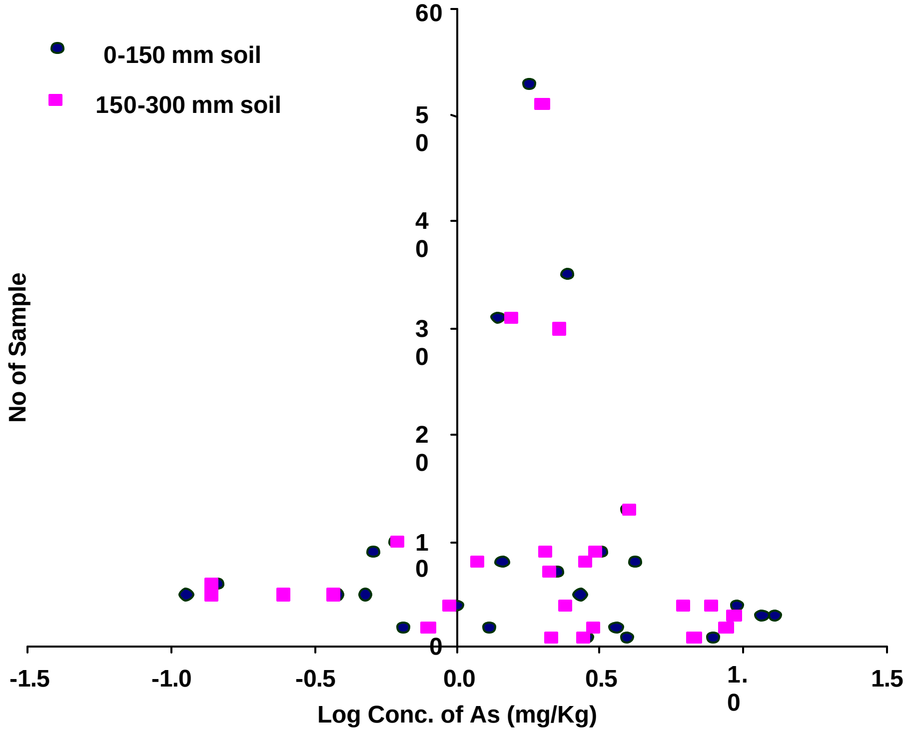

Figure 5: Log distribution of arsenic concentration in soil samples

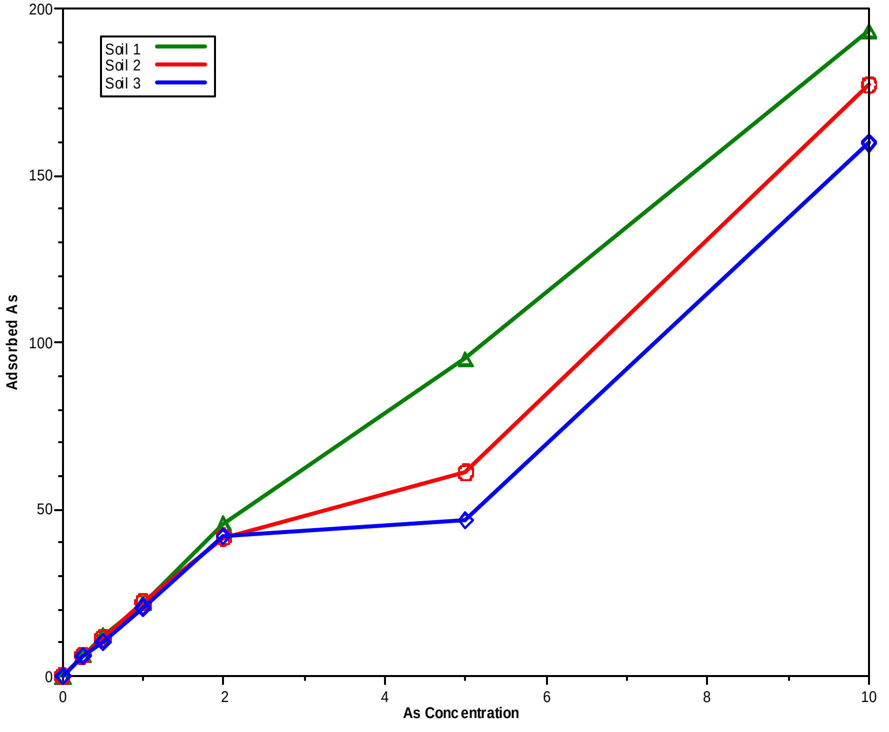

Figure 6: Adsorption of arsenic (mg/kg) in soil as affected by clay contents and As concentration (mg/L) in soil solution. Soil-1 (>84% clay); Soil-2 (>55% clay); Soil-3 (<20% clay)

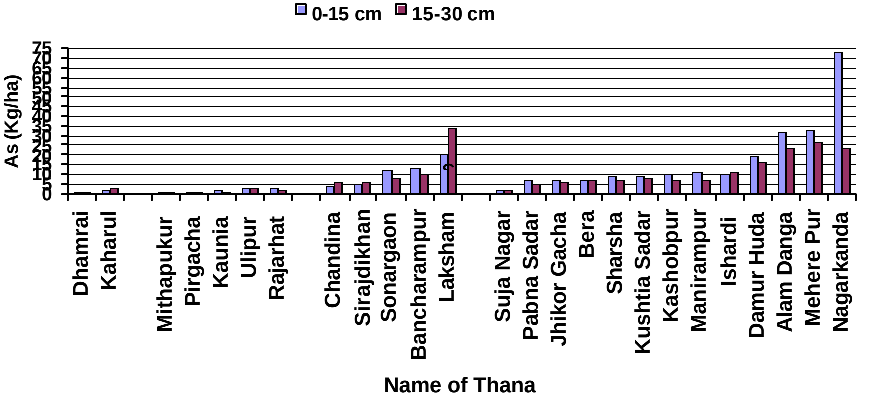

Figure 7: Arsenic amount in soils of different thanas

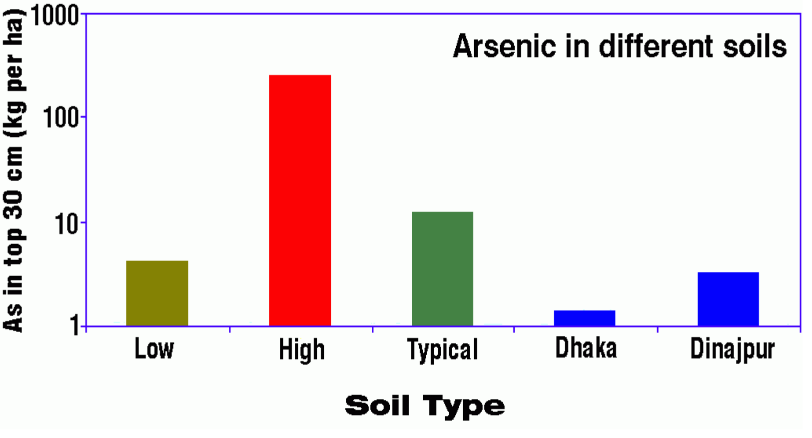

Figure 8: Average As content in contaminated soil and uncontaminated soils (Dhaka and Dinajpur)

Arsenic Loading in soils

Table 2 shows values of As in water and the corresponding values of As in soils. It may be mentioned here that all these water values are not necessarily irrigation water values.

The As concentration in water used for irrigation varied between 0.136 to 0.55 mg L^{-1}. Using these values, total As loading in irrigated soils for a Boro rice that requires 1000 mm of irrigation water per season, ranges from between 1.36 to 5.5 kg ha^{-1} yr^{-1}. Similarly, for winter wheat that requires 150 mm of irrigation water per season As loading from irrigation ranges from between 0.12 to 0.82 kg ha^{-1} yr^{-1}. The As loads for other crops requiring irrigation are also presented in Figures 9 and 10. Based on As loading from irrigation, and the average yield per hectare of some commonly consumed crops, the soil build-up of As has been calculated (Fig. 11). It is apparent from Figure-11 that the build up of As in surface through irrigation water, though not very high, is the greatest for Arum (*Colocassia antiquorum*) followed by Boro rice that requires supplemental irrigation.

When the water extractable As was determined, it was found that only in a few soils, there was water soluble fraction. Where this fraction could be determined, the proportion of water extractable fraction to aqua-regia fraction (total) was quite negligible (Table 3). Moreover, the correlation between total As and water extractable As is poorly correlated (r =

0.00057). This gives an indication that soil build-up of As from ground water irrigation is likely to be soil dependent phenomenon, not a generalized one. The corollary to this assumption is that, depending on the soil type, As in ground water will enter freely into the growing crop.

Table 2: Arsenic in water and corresponding As in soils

Location	Water As (mg/kg)	Soil Depth,cm	Soil As (mg/kg)
Sharsha	0.041	0-15	13.670
Sirjdikhan	0.544	15-30	10.655
Alamdanga	0.021	0-15	16.647
Alamdanga	0.021	15-30	11.820
Alamdanga	0.191	0-15	11.918
Alamdanga	0.058	0-15	10.675
Meherepur	0.163	0-15	33.912
Meherepur	0.016	15-30	28.220
Laksham	0.145	15-30	10.791
Laksham	0.658	15-30	39.107
Laksham	0.729	0-15	18.125
Laksham	0.037	15-30	16.971
Laksham	0.261	0-15	28.009
Laksham	0.261	15-30	42.608
Laksham	0.397	0-15	22.763
Laksham	0.341	15-30	12.529
CHANDINA	0.380	15-30	19.270
CHANDINA	0.160	15-30	19.270
Sonargaon	0.682	0-15	38.930
Sonargaon	0.860	0-15	22.866
Sonargaon	0.860	15-30	14.829
Sonargaon	0.860	0-15	14.000
Sonargaon	0.860	15-30	13.671
Bancharampur	0.092	0-15	17.147
Bancharampur	0.115	0-15	11.318
Nagarkanda	0.077	0-15	81.248
Nagarkanda	0.064	15-30	26.559

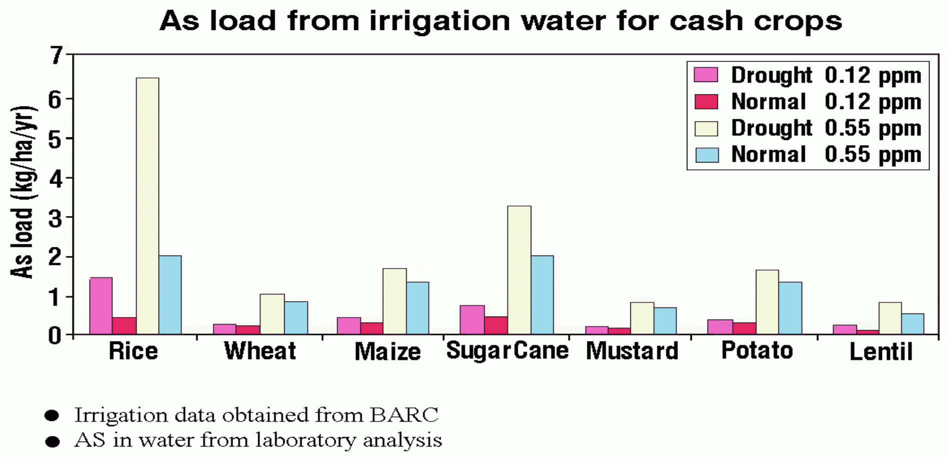

Figure 9: Arsenic load in soils for some cash crops

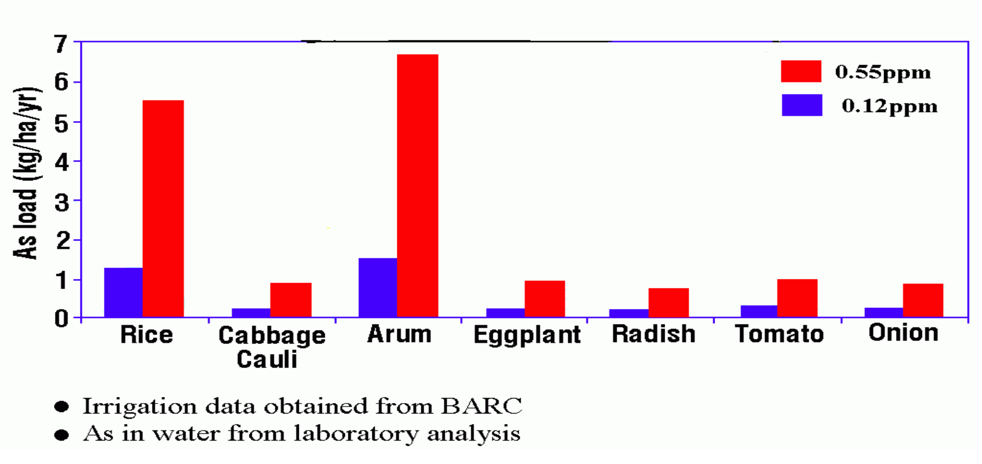

Figure 10: Arsenic load from irrigation water for some vegetables as compared that for rice

Other soil parameters like pH, EC, P, Fe etc. were determined to find any relationship with As . It was observed that a positive correlation existed between total soil As and soil pH. Negative correlation was obtained with total soil As and soil EC, soil P, soil S, soil K and water soluble Fe (Table 4). The relationship between water extractable As and water extractable Fe is also shown in Table 4.

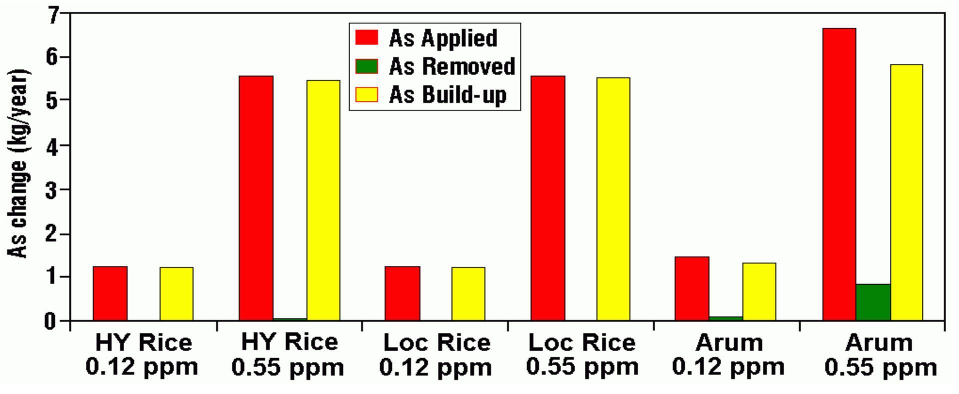

Figure 11: Arsenic budget for water-soil-crop transfer

Estimation based on laboratory values and data obtained from BARC:
- Irrigation water had 0.12 or 0.55 mg L^{-1} As
- Plant As content data are based on moderate and extreme cases

Irrigation rates for rice and arum taken as 1 m and 1.2 m per crop, respectively.

Table 3: Total As content and corresponding water extractable As content in some soils

Location	Depth (in mm)	Total As (mg/Kg)	Water extractable As (mg/Kg)
Sirajdikhan	0-150	4.930	0.041
	150-300	3.597	0.022
Sirajdikhan	0-150	5.634	0.012
	150-300	1.296	0.013
Chandina	0-150	3.321	0.050
	150-300	1.344	0.012
Chandina	0-150	2.044	0.014
	150-300	0.593	0.017
Sonargaon	0-150	9.915	0.033
	150-300	0.317	0.027
Kushtia	150-300	2.223	0.713
Dhamarai	0-150	0.327	0.06
Jessore	0-150	13.67	0.21
Jessore	0-150	1.6	0.21
Jessore	0-150	5.84	0.026

Table 4: Correlation coefficient values between soil As and other parameters

Correlation value soil pH and Soil As	0.14
Correlation value soil EC and Soil As	-0.03
Correlation value soil P and Soil As	-0.10
Correlation value soil S and Soil As	-0.26
Correlation value soil K and Soil As	-0.29
Correlation value soil Fe and soil As	-0.18
Correlation value w.e.Fe and w.e.As	-0.11

CONCLUSION

It is apparent from the findings of the present work that the average As content in Bangladesh soils are less than 10 mg/kg. The surface 150 mm soil contain more As than the subsurface 300 mm soil. The background value of soil As is much below the average in areas where ground water contamination has not been reported or where the ground water is very feebly contaminated. There is no direct relationship with As in ground water and corresponding As in soil. However, there is a tendency of soil build-up of As in some cases where As-contaminated ground water is used for irrigation. The release of soil As has no bearing on the total As content of soils. There is a positive relationship of As adsorption in soils with its clay content. pH of soils seems to play certain role in this regard.

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